Paper

ASSESSMENT OF PLUTONIUM EXPOSURE IN THE ENEWETAK POPULATION BY URINALYSIS

L. C. Sun, C. B. Meinhold, A. R. Moorthy, E. Kaplan, and J. W. Baum*

Abstract—**Since 1980, the inhabitants of Enewetak Atoll have been monitored periodically by scientists from Brookhaven National Laboratory for internally deposited radioactive material. In 1989, the establishment of fission track analysis and of a protocol for shipboard collection of 24-h urine samples significantly improved our ability to assess the internal uptake of plutonium. The purpose of this report is to show the distribution of plutonium concentrations in urine collected in 1989 and 1991, and to assess the associated committed effective doses for the Enewetak population based on a long-term chronic uptake of low-level plutonium. To estimate dose, we derived the plutonium dose-per-unit-uptake coefficients based on the dosimetric system of the International Commission on Radiological Protection. Assuming a continuous uptake, an integrated Jones's plutonium urine excretion function was developed to interpret the Enewetak urine data. The Appendix shows how these values were derived. The committed effective doses were 0.2 mSv, calculated from the 1991 average plutonium content in 69 urine samples.**
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Key words: Marshall Islands; plutonium; excretion, urinary; dose assessment

INTRODUCTION

ENEWETAK ATOLL, in the Republic of the Marshall Islands (RMI), lies within the former Pacific Proving Grounds and was chosen as the site for nuclear testing from 1948 to 1951 because of its remoteness location and its geological features. In December 1947, the United States relocated all 136 residents of Enewetak to Ujeland Atoll before starting a program of nuclear-weapons tests. Forty-three tests were conducted between 1948 and 1951 resulting in radiological contamination of the atoll. Plutonium activities measured in the top 2 cm of the soil ranged from 0.4 to 17 mBq g^{-1} with a median of 4.4 on Enewetak Island (Wilson et al. 1975). From 1972 to 1978, major efforts were made to remove the top 30 cm of soil from the Island; then, the soil was buried on Runit Island, located at the northern Enewetak Atoll. The cleanup guidelines were (1) the soil should be removed if the plutonium concentration exceeded 15 Bq g^{-1} (400 pCi g^{-1}); (2) soil could be left in place if the concentration was less than 1.5 Bq g^{-1} (40 pCi g^{-1}); and (3) for concentrations ranging between 1.5–15 Bq g^{-1}, a decision should be made on a case-by-case basis (DNA 1981). Repatriation of the Enewetak population was begun immediately after the cleanup programs. Now, the largest inventory of plutonium on Enewetak Atoll remains in the sediments of the lagoon (Wilson et al. 1975; Nevissi and Schell 1975; Robison et al. 1978, 1980, 1987).

Since 1957, members of the Marshall Islands Program at Brookhaven National Laboratory (BNL) have routinely visited RMI to assess the acceptability of the Enewetak population living on the island by determining their internally deposited radionuclides using whole-body counting and urinalysis methods (Greenhouse et al. 1980; Miltenberger et al. 1981; Lessard et al. 1984; Conard 1992; Sun et al. 1992, 1995). The purposes of this report are to describe the new protocol established for collecting urine samples in 1989 and 1991, to show the distribution of plutonium concentrations using fission track analysis (FTA) urinalysis, and to assess the associated committed effective doses for people of Enewetak, based on a continuous, long-term chronic uptake of plutonium. The FTA method was developed at BNL for analyzing low levels of plutonium in urine (Moorthy et al. 1988). All plutonium data discussed in this paper were analyzed by the FTA method at BNL.

MATERIALS AND METHODS

Methods for interpreting urine data and estimating plutonium dose

The International Commission on Radiological Protection (ICRP) Publication 56 (1990) provides the age-dependent dose-coefficient factors (DCF) for computing internal dose (Leggett 1984, 1985). Due to the long half-life of ^{239}Pu and its lengthy retention in the body, the age-dependent ingestion dose-coefficients after 1 y to adulthood show only small variation. They can be rounded to about 1.0×10^{-6} Sv Bq^{-1}, with a gastrointestinal tract absorption f_1 value of 10^{-3}. The calculated uptake dose coefficient is 1 mSv Bq^{-1} (10^{-6} Sv $Bq^{-1} \div f_1$).

Table 1 shows the calculated committed effective dose-coefficients due to uptake based on recommenda-

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Table 1. Comparison of the ICRP recommended plutonium dose coefficients (mSv Bq^{-1}) for uptake.

Age-specific groups	ICRP Publication 56 (1990)	ICRP Publication 67 (1993)	Constant ratio ICRP 67 ÷ ICRP 56
3 mo	1.4	0.84	0.60
1 y	1.4	0.84	0.60
5 y	1.1	0.66	0.60
10 y	1.0	0.54	0.54
15 y	0.98	0.50	0.51
Adult (18 y and older)	0.97	0.50	0.52

tions in ICRP Publications 56 (1990) and 67 (1993), for calculating dose. The results were obtained by using the individual ingestion pathway dose coefficients divided by the recommended f_1 values. ICRP revised its recommendations for calculating plutonium dose from those given in Publication 56 to new values in Publication 67 because of (1) the f_1 value (age 1 y to 70 y) was reduced by a factor of two (from 1×10^{-3} to 5×10^{-4}), and (2) the tissue weighing factor (w_T) of the bone surface was reduced by a factor of three (from 0.03 to 0.01) in ICRP Publication 60 (1991). The last column of Table 1 shows the ratio of the values from Publication 67 over those of Publication 56; it indicates that the plutonium hazard in the body may be overestimated, and the revised age-specific dose-coefficients from the latest ICRP recommendations are just above one half of the committed effective dose values given in Publication 56.

To interpret 24-h urine data, both Durbin's (1972) and Jones's (1985) plutonium urinary excretion function are the ones most accepted (ICRP 1988; Lessard et al. 1987). Durbin's predicted elimination rate is much faster than that of Jones. By using a model with a fast elimination rate like that of Durbin, an enormous overestimation of plutonium body content can result from long-term post-exposure urine measurements. Therefore, the Jones function is chosen for this study; its predicted values (fraction per unit single acute uptake) for a 100-d and 10,000-d post-uptake are $\sim10^{-4}$ and $\sim10^{-5}$, respectively. Therefore, detection of 1 μBq of ^{239}Pu can be interpreted as a committed effective dose as low as 10μ Sv via a 24-h urine sample collected 100 d after an acute uptake (i.e., 1 mSv Bq^{-1} × 1 μBq d^{-1} ÷ 10^{-4} d^{-1}). Similarly, detection of 1 μBq of ^{239}Pu also can be interpreted as a committed effective dose of 0.1 mSv to age 70 y after 10,000 d (~30 y) of such an uptake (i.e., 1 mSv Bq^{-1} × 1 μBq d^{-1} ÷ 10^{-5} d^{-1}). However, the accuracy of assessment of the dose would be best if the plutonium entered in the body by an injection, the physical size of the particles was 1 activity median aerodynamic diameter (AMAD), and its chemical solubility was in Lung-Class Y (ICRP 1984).

Method of calculating dose from Enewetak urine samples

Since the people of Enewetak were repatriated in May 1980, we assumed for this study that all plutonium intake occurred after they returned to live in their Enewetak homeland. Urine samples were collected in June 1989 and February 1991. Therefore, the ten years from 1980 to 1990 gave us a convenient number for assessing the fraction of the total plutonium uptake over this period that would be eliminated by the last day of the period. Only a few methods are available for interpreting plutonium urine data due to chronic exposure (Butler 1972; Ramsdem et al. 1990). In general, methods of solving convolution integrals of an excretion function were suggested for assessing internal uptake resulting from recurrent or prolonged exposure (ICRP 1969).

Based on Jones's (1985) urinary functions, an integrated procedure was developed for interpreting 24-h urine elimination to assess plutonium body burdens under conditions of constant, continuous, chronic uptake. Both Jones's (1985) function and the integrated arithmetics are described in the Appendix. The calculated integrated elimination rate of plutonium in a 24-h urine sample after a 10-y constant chronic uptake is 2.4×10^{-5}. This constant means that the measured plutonium activity as a 24-h total represents 0.0024% of that in the body. Similarly, the calculated 24-h urine elimination rates of plutonium at the end of 9 and 11 y are 2.5×10^{-5} and 2.3×10^{-5}, respectively, under the same conditions of uptake. Because of the slow rate of elimination of plutonium from the body, the 9-y, 10-y, and 11-y urinary elimination fractions into a 24-h urine sample do not differ by more than 5% within an interval of 1 y. Therefore, when a DCF of 1 mSv Bq^{-1} is used, each 1 μBq d^{-1} of ^{239}Pu indicates a 0.04 mSv committed effective dose (i.e., 1 mSv Bq^{-1} × 1 μBq d^{-1} ÷ 2.4×10^{-5} d^{-1}) to age 70 y for the people of Enewetak.

Urine sample collection protocol

Before 1989, urine bottles (Nalgene™,† 2-L high-density polyethylene) were distributed directly to volunteers who collected the samples at home and then placed their bottles in collection boxes at the seashore to be picked up the next day. This protocol gave no assurance that dust and sand was kept out of the bottles, nor was there any guarantee that the urine in the bottle was from the donor named on the outside; further, there was no provision for insuring that the sample represented a 24-h elimination.

In 1989, a shipboard 24-h urine sample collection protocol was developed for the Marshallese (Sun et al. 1993). This protocol was designed to minimize the unwanted contamination and to assure the quality of all 24-h urine samples on ships. The following steps were instituted: (1) all collection bottles were controlled and handled on board by registered nurses and authorized staff; (2) all the Marshallese participants showered and changed into clean clothes provided by BNL staff and stayed on board for the entire interval; (3) a collection log (e.g., name, date of birth, sex, photo ID, urine volume, and elimination time) was properly completed; and (4) all samples were acidified at the end of the 24-h

†Nagle Nune International, 75 Panorama Creek Drive, Rochester, NY 14602

collection to minimize plutonium plating on the inner surface of the collection bottle. All samples were collected under uniform, controlled and monitored environments to reduce unwanted contamination and to insure a full 24-h collection.

Table 2 compares the plutonium contents determined from 24-h eliminations of 32 Marshallese (in Column 1) for whom samples had been obtained under both the old on-shore and the 1989 shipboard protocols. Column two shows the range of ^{239}Pu activity in 24-h urine, from 10 to 297 μBq. These samples were collected between 1981 and 1984 using the on-shore collection protocol described above. Column three shows that only 2 samples (from Subject No. 1 and No. 3) were just above the MDL (defined at 99% confidence level) of 2 μBq of ^{239}Pu using the shipboard protocol in 1989. The difference in the paired plutonium contents between Column 2 and Column 3 can only be explained as due to external contamination of the samples during on-shore collections. Hence, Table 2 shows the benefit of the shipboard protocol and the superior quality of the 1989 urine samples. A disadvantage of the shipboard protocol is that it requires an overnight stay on the ship, and, therefore, participation is limited to those people who do not have pressing domestic responsibilities; this group generally consists of people between 8–18 y old.

Table 2. Comparison of ^{239}Pu content (μBq) in the urine of 32 Marshallese under two collection protocols.

Subject No.	On-shore protocol (1981–1984)	Shipboard protocol (1989)
1	297	2.75
2	175	<2
3	127	2.75
4	125	<2
5	125	<2
6	125	<2
7	119	<2
8	119	<2
9	99.5	<2
10	99.5	<2
11	89.7	<2
12	86.4	<2
13	85.0	<2
14	77.3	<2
15	71.7	<2
16	68.6	<2
17	61.5	<2
18	60.3	<2
19	53.7	<2
20	51.3	<2
21	48.4	<2
22	47.9	<2
23	42.6	<2
24	42.4	<2
25	39.0	<2
26	23.6	<2
27	20.3	<2
28	16.6	<2
29	16.6	<2
30	12.3	<2
31	11.7	<2
32	10.4	<2

Information on 1989 and 1991 urine samples

Due to limited facilities on the ship (e.g., beds, toilets and shower rooms) it was inconvenient to collect urine from more than 10–12 volunteers per day. Therefore, groups of males or females participated on alternate days. On each trip there was a maximum of 80 people tested because we were also limited by the amount of food and tap-water available on the vessel. Seventy-two and sixty-nine FTA were determined for the people from Enewetak in 1989 and 1991, respectively. In 1989, there were 39 male and 33 female volunteers; most of them were teenagers. The average urine volumes of these males and females were 830 and 820 mL, respectively. In the past, we found that the range of the 24-h urine volume from the Marshallese collected using shipboard protocol was 50 to 3,500 mL (Sun et al. 1993). For dose assessment, we decided to use data only for individuals with volumes greater than 300 mL because smaller samples might not constitute a normal 24-h excretion.

URINE RESULTS

Table 3 shows the distribution of plutonium contents in the 24-h urine samples. The ^{239}Pu values in the samples were arbitrarily subdivided into eight groups, ranging from less than 1 μBq to greater than 37 μBq (Column 1). The MDL associated with these samples was 2 and 3 μBq, for 1989 and 1991, respectively (Sun et al. 1995). The statistical parameters shown at the bottom of Table 3 are the total number of samples (n), the samples' mean ($\bar{x}$), and the samples' standard deviation (s). The calculated $\bar{x}$ and s values were based on the net activities, even though some were negative or measured zero.

1989 FTA results

In Column 2 of Table 3, data from 72 people show that ~90% of the 24-h samples had below 3 μBq d^{-1}, and none of them was equal to or higher than 7 μBq d^{-1}. The committed effective dose to age 70 y for a 7 μBq 24-h urine sample was estimated to be about 0.3 mSv.

Table 3. Distribution of ^{239}Pu activity (μBq d^{-1}) and the corresponding values of statistical parameters (n, $\bar{x}$, s) for the people of Enewetak.

^{239}Pu activity in 24-h urine sample (μBq)	1989	1991
	Frequency distribution	
$x < 1$	41	17
$1 \le x < 3$	23	23
$3 \le x < 5$	5	18
$5 \le x < 7$	3	4
$7 \le x < 9$	0	5
$9 \le x < 11$	0	1
$11 \le x < 37$	0	0
$x \ge 37$	0	1[a]
n =	72	69
$\bar{x}$ (μBq) =	1.0	4.7
s (μBq) =	1.7	17

[a] This individual value was 146 μBq.

The $\bar{x}$ and s were 1.0 and 1.7 μBq, respectively. The CV value was 170%, where CV (%) = 100 × ($s/\bar{x}$). Therefore, the calculated effective dose to age 70 y due to ^{239}Pu is 0.04 mSv (i.e., 1 mSv Bq^{-1}×1 μBq d^{-1} ÷ 2.4×10^{-5} d^{-1}) for an average person living on Enewetak.

1991 FTA results

In Column 3 of Table 3, values from 69 people show that ~40% were above the MDL value of 3 μBq. The $\bar{x}$ and s of the 1991 samples were 4.7 μBq d^{-1} and 17 μBq d^{-1}, respectively. The committed effective dose was about 0.2 mSv (i.e., 1 mSv Bq^{-1} × 4.7 μBq d^{-1} ÷ 2.4×10^{-5} d^{-1}). The large standard deviation value is due to the 146 μBq in the data set. The $\bar{x}$ and s recalculated without this value are 2.6 and 2.2, respectively, and the CV values fall from 362% to 85%. In this case, the estimated effective dose is ~ 0.1 mSv. Individual urinary excretion data can be highly variable (Clemente and Delle Site 1982; ICRP 1988), and presently it is difficult to identify the cause of the higher ^{239}Pu average in the 1991 results. More urine samples are to be collected from the individuals to evaluate this suspect issue.

DISCUSSION AND CONCLUSION

In Publication 54 (1988), the ICRP indicated that monitoring urine for intakes of plutonium, especially for low levels, can present difficulties in making the measurements and in interpretation. The ICRP recommended using a series of excretion measurements to evaluate an individual intake (ICRP 1988). For monitoring or measuring low-level plutonium in an individual, a repetitive series of urine measurements is required. Furthermore, for the Enewetak population, environmental measurements, analyses of environment pathways, and dietary studies also can be used to estimate intake and corroborate the results of urine bioassay to enhance reliability of the dose assessment.

The algorithm described in the Appendix was specifically developed to interpret urine data for assessing plutonium uptake in the people of Enewetak. The main limitation of this method is the requirement that the intake rate is a constant steady state during the integrated time interval. If the recurrent intake rates were unevenly distributed over this period, then the dose will be underestimated if more plutonium was taken up within the early half of the integrated time. On the other hand, the dose will be overestimated if such intakes were greater within the later half (recent) of the time. In either case, the uncertainty is within a factor of 2.4 (i.e., 2.4×10^{-5} ÷ 1.0×10^{-5}), estimated from the Appendix.

Based on both the Jones (1985) urine excretion function and the ICRP Publication 56 (1990) systemic retention model for plutonium, and also upon the average ^{239}Pu content in the 1991 urine samples, we calculate an average ^{239}Pu uptake of 4.7 Bq and a committed effective dose of 0.2 mSv to age 70 y for an average adult. This calculation assumed a constant chronic intake and included the 146 μBq d^{-1} datum point; its inclusion is appropriately conservative. The estimated average dose for the people of Enewetak would be reduced by about one half if the DCFs of ICRP Publication 67 (1993) were used.

Again, the dose assessed is a population average, and the uncertainty, in percent, can be measured from the mean associated CV. More samples were collected after 1991 by BNL staff for plutonium urinalysis to establish individual plutonium exposure records after 1991. Although the committed effective dose to the Enewetak inhabitants from plutonium is low, plutonium is perceived by Enewetak people as being the element of utmost concern in their environment.

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REFERENCES

Butler, G. C. Retention and excretion equations from different patterns of uptake. In: Assessment of Radioactive Contamination in Man. Vienna: IAEA; IAEA-SM-150/42; 1972: 495–508.

Clemente, G. H.; Delle Site, A. Assessment of plutonium internal contamination in man. Luxembourg: Commission of the European Communities; EUR-7157-EN; 1982.

Conard, R. A. Fallout: The experiences of a medical team in the care of a Marshallese population accidentally exposed to fallout radiation. Upton, NY: Brookhaven National Laboratory; Report BNL-46444; 1992.

Defense Nuclear Agency. The radiological cleanup of Enewetak Atoll. Washington, DC: DNA; 1981.

Durbin, P. W. Plutonium in man: A new look at the old data. In: Stover; Jee, eds. The radiobiology of plutonium. Salt Lake City: J. W. Press; 1972: 469–530.

Greenhouse, N. A.; Miltenberger, R. P.; Lessard, E. T. Dosimetric results for the Bikini population. Health Phys. 38:845–851; 1980.

International Commission on Radiological Protection. The assessment of internal contamination resulting from recurrent or prolonged uptakes. Oxford: Pergamon Press: ICRP Publication 10A; 1969.

International Commission on Radiological Protection. Individual monitoring for intake of radionuclides by workers: Design and interpretation. Oxford: Pergamon Press; ICRP Publication 54; Annals of the ICRP 19 (1–3); 1988.

International Commission on Radiological Protection. Age-dependent doses to members of the public from intake of radionuclides. Oxford: Pergamon Press; ICRP Publication 56, Part 1; Annals of the ICRP 20(2); 1990.

International Commission on Radiological Protection. 1990 recommendations of International Commission on Radiological Protection. Oxford: Pergamon Press; ICRP Publication 60; Annals of the ICRP 21(1); 1991.

International Commission on Radiological Protection. Age-dependent doses to members of the public from intake of radionuclides. Oxford: Pergamon Press; ICRP Publication 67, Part 2; Annals of the ICRP 23(3/4); 1993.

Jones, S. R. Derivation and validation of a urinary excretion function for plutonium applicable over tens of years post uptake. Radiat. Protect. Dosim. 11:19–27; 1985.

Leggett, R. W. Bioassay data and a retention-excretion model for systemic plutonium. Oak Ridge, TN: Oak Ridge National Laboratory; NUREG/CR-3346; 1984.

Leggett, R. W. A model of the retention, translocation and excretion of systemic Pu. Health Phys. 49:1115–1137; 1985.

Lessard, E. T.; Miltenberger, R. P.; Cohn, S. H.; Masolino, S. V.; Conard, R. A. Protracted exposure to fallout: The Rongelap and Utirik experience. Health Phys. 46:511–527; 1984.

Lessard, E. T.; Xia, Y.; Skrable, K. W.; Chabot, G. E.; French, C. S.; Labone, T. R.; Johnson, J. R.; Fisher, D. R.; Belanger, R.; Lipsstein, J. L. Interpretation of bioassay measurements. Washington, DC: US. Nuclear Regulatory Commission; NUREG/CR-4884; 1987

Marsh, K. V.; Jokela, T. A.; Eagle, R. J.; Noshkin, V. E. Radiological and chemical studies of ground water at Enewetak Atoll, 2. Residence time of water in cactus crater. Livermore, CA: Lawrence Livermore National Laboratory; UCRL-51913 Pt. 2; 1978.

Miltenberger, R. P.; Lessard, E. T.; Greenhouse, N. A. Cobalt-60 and cesium-137 long-term biological removal rate constants for the Marshallese population. Health Phys. 40:615–623; 1981

Moorthy, A. R.; Schopfer, C. J.; Banerjee, S. Plutonium from atmospheric weapons testing: Fission track analysis of urine samples. Analytical Chem. 60:857A; 1988.

Nevissi, A.; Schell, W. R. ^{210}Po and ^{239}Pu, ^{240}Pu in biological and water samples from the Bikini and Enewetak Atolls. Nature 225:321; 1975.

Noshkin, V. E.; Wong, K. M.; Marsh, K.; Eagle, R.; Holladay, G. Plutonium radionuclides in the groundwater at Enewetak Atoll. In: Proceedings of IAEA Symposium on Transuranium Nuclides in the Environment. Vienna, Austria: IAEA; SM-199/33; 1976: 517–543.

Ramsdem, D.; Birchall, A.; Bull, R. K.; Foster, P. P.; Gibson, J. A. B.; Strong, R.; Taylor, N. A.; Tyler, G. R.; Wraight, J. Laboratory intercomparison of methods used for the assessment of systemic burdens of plutonium. Radiat. Protect. Dosim. 30:95–99; 1990.

Robison, W. L.; Noshkin, V. E.; Phillips, W. A. Assessment of potential doses to populations from the transuranic radionuclides at Enewetak Atoll. Livermore, CA: Lawrence Livermore Laboratory; Report UCRL-52666; 1978.

Robison, W. L.; Phillips, W. A.; Mount, M. E.; Clegg, B. R.; Conrado, C. L. Reassessment of the potential radiological doses for residents resettling Enewetak Atoll. Livermore, CA: Lawrence Livermore Laboratory; Report UCRL-53066; 1980.

Robison, W. L.; Conrado, C. L.; Phillips, W. A. Enjebi Island dose assessment. Livermore, CA: Lawrence Livermore National Laboratory; Report UCRL-53805; 1987.

Sun, L. C.; Meinhold, C. B.; Moorthy, A. R.; Clinton, J. H.; Kaplan, E. Radiological dose assessments in the Northern Marshall Islands (1989–1991). In: Proceedings of the Eighth International Congress of the International Radiation Protection Association (IRPA-8). Montreal: IRPA; Vol. II. BNL-45868; 1992: 1320–1323.

Sun, L. C.; Clinton, J. H.; McDonald, J.; Moorthy, A. R.; Kaplan, E.; Meinhold, C. B. Urine collection protocol in the Republic of the Marshall Islands. Radiat. Protect. Management 10:64–72; 1993.

Sun, L. C.; Moorthy, A. R.; Kaplan, E.; Baum, J. W.; Meinhold, C. B. Assessment of plutonium exposures in Rongelap and Utirik populations by fission tracks analysis of urine. Applied Radiat. Isotopes 46:1259–1269; 1995.

Wilson, D. W.; NG, Y. C.; Robison, W. L. Evaluation on plutonium at the Enewetak Atoll. Health Phys. 29:599–611; 1975.

APPENDIX

This procedure is used to interpret the significance of levels of plutonium found in a urine sample obtained after a constant, continuous, chronic uptake. Let $I(t)$ be the uptake rate in day t, and $J(t)$ be the urine fraction in t day using Jones's (1985) plutonium urine excretion function, as in the following:

$$J(t) = 0.00475e^{-0.558t} + 0.000239e^{-0.0442t} + 0.0000855e^{-0.00380t} + 0.0000142e^{-0.0000284t}, \quad \text{(A1)}$$

where all the eigenvalues associated with exponential terms have a unit of reciprocal days. Then, the fraction of plutonium excreted after the first 24-h of uptake is

$$P(1) = I(1) \times J(1).$$

Similarly, plutonium excreted after each 24-h (day) is

$$P(2) = I(1) \times J(2) + I(2) \times J(1); \quad \text{(A2)}$$

$$P(3) = I(1) \times J(3) + I(2) \times J(2) + I(3) \times J(1); \quad \text{(A3)}$$

$$P(4) = I(1) \times J(4) + I(2) \times J(3) + I(3) \times J(2) + I(4) \times J(1); \quad \text{and so forth.} \quad \text{(A4)}$$

Therefore,

$$P(n) = I(1) \times J(n) + I(2) \times J(n-1) + \ldots + I(n) \times J(1). \quad \text{(A5)}$$

If

$$I = I(1) = I(2) = I(3) = I(4) = \ldots = I(n) \quad \text{(A6)}$$

is a constant continuous uptake, then

$$P(n) = I \times [J(1) + J(2) + J(3) + \ldots + J(n-1) + J(n)] = I \times \sum_{i=1}^{n} J_i(t) = I \times n \times \frac{\int_0^n J(t)\,dt}{n}, \quad \text{(A7)}$$

where $P(n)$ is the n^{th} day plutonium excretion in 24-h and "$I \times n$" is the sum of total uptakes from days 1 to n.

Hence,

$$\eta(n) = \frac{P(n)}{I \times n} = \frac{\int_0^n J(t)\, dt}{n}. \qquad \text{(A8)}$$

So, $\eta(n)$ is the fraction of the total uptake of plutonium to be excreted in the n^{th} day 24-h urine sample that equals the integration sum of Jones's function from 0 to n days and divided by n. Using Jones's plutonium urine excretion function, we calculate $\eta(3{,}650) = 2.4 \times 10^{-5}$.

■■